First Demonstration of Pump and Probe Experiment for Wide-gap Semiconductors Using Free Electron Laser and Synchronously-operated Femtosecond Laser

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Outline

- XFEL Project in the world
- Necessity for fast scintillator
- Hydrothermal method-grown ZnO crystal as scintillator
- Improvement of ZnO scintillator by impurity doping
- Pump and probe experiments using ZnO crystal
- Summary

XFEL projects

DESY in Europe

SACLA in Japan

SLAC in USA

New scientific fields have already opened.

Newest X-ray Free Electron Laser (XFEL) Facility

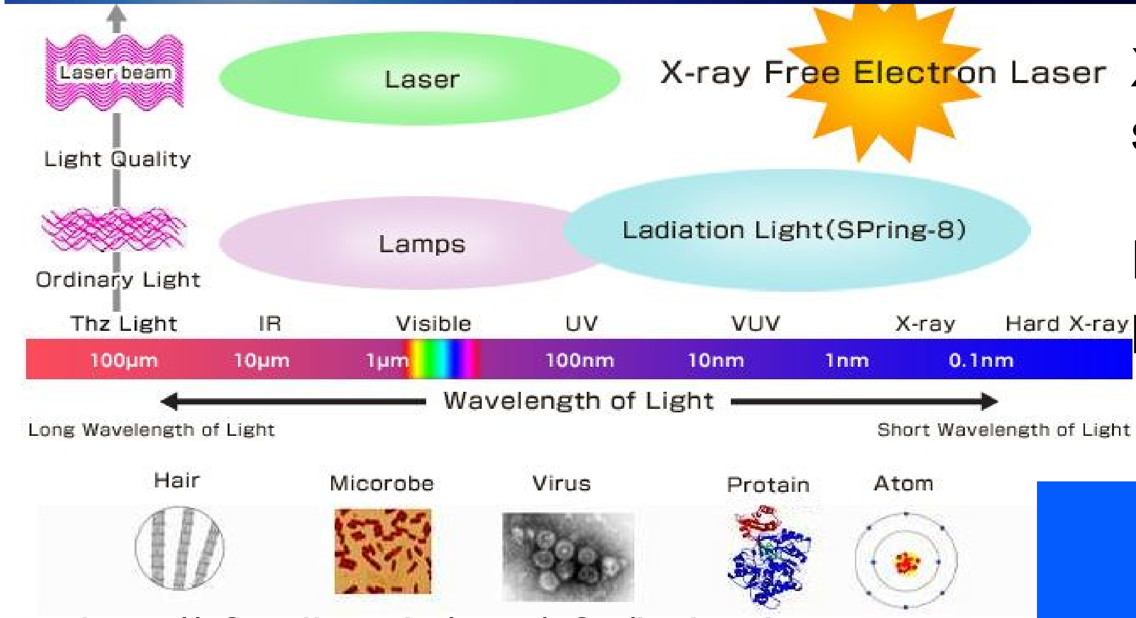
June 7, 2011: λ = 0.12 nm

June 10, 2011: $\lambda = 0.1$ nm → Shortest in the world

Expected peak brightness: 1×10^8 times compared to SPring8

Expected $\Delta\tau$: <10 fs

X-ray structure analysis by XFEL



XFEL has characteristics of lasers and synchrotron radiations.

Femtosecond pulse duration and high peak brightness in the X-ray region.

http://xfel.riken.jp/eng/xfel/index.html

- Structural analysis of a protein molecule
- Direct observation of atomic motion during photo chemical reaction
- Research of new materials

http://zms.desy.de/index_eng.html

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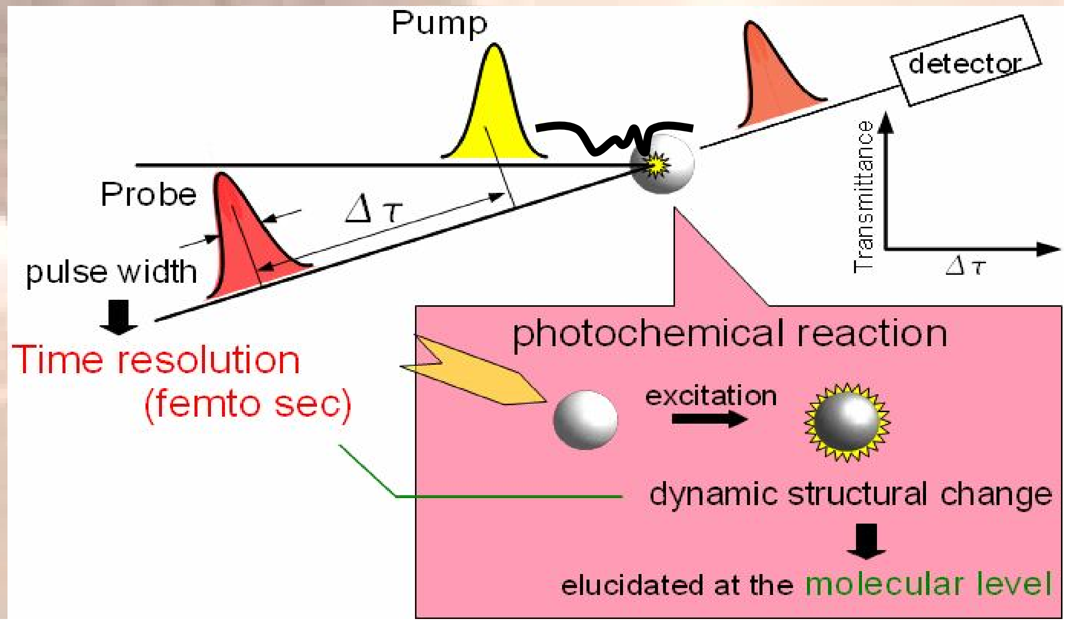
Why is a fast scintillator needed for XFEL facilities?

High accuracy temporal overlap with optical pulse is required for pump and probe experiments

FEL and optical pulse should be synchronized within 0.1 psec accuracy.

The response time of conventional scintillator (CeYAG) is about 50 ns



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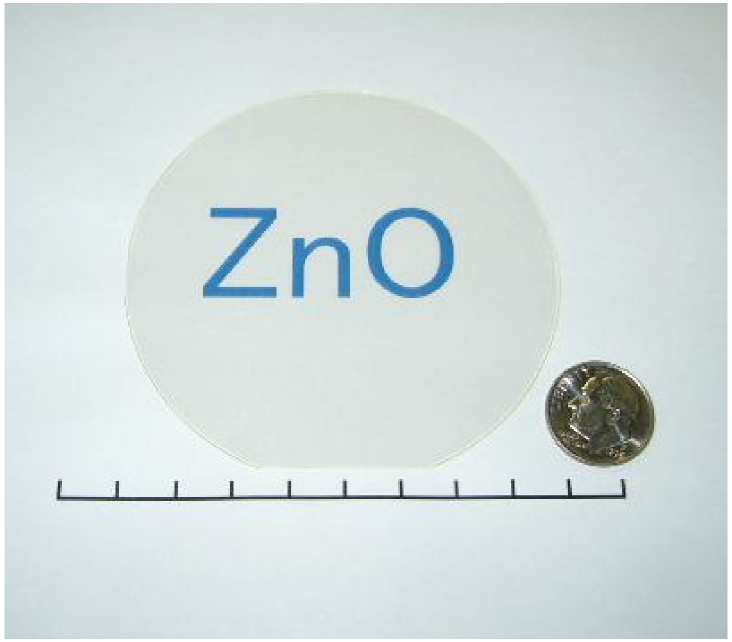
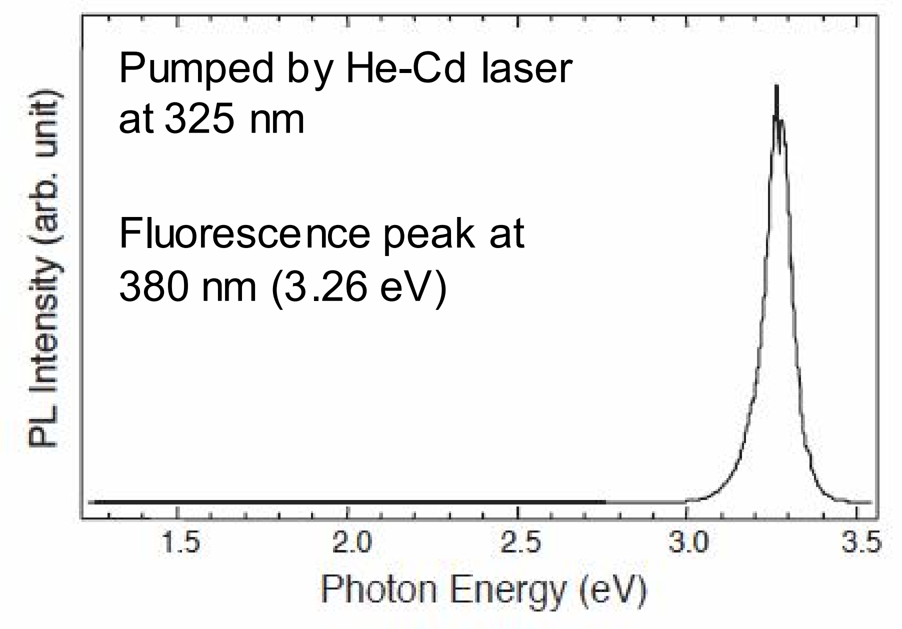
ZnO crystal grown by the Hydrothermal method

Short fluorescence decay time of ~1 ns.
Useful emission wavelength (Transparent for glass).
Large-sized single crystal of up to 3 inch-diameter can be grown.

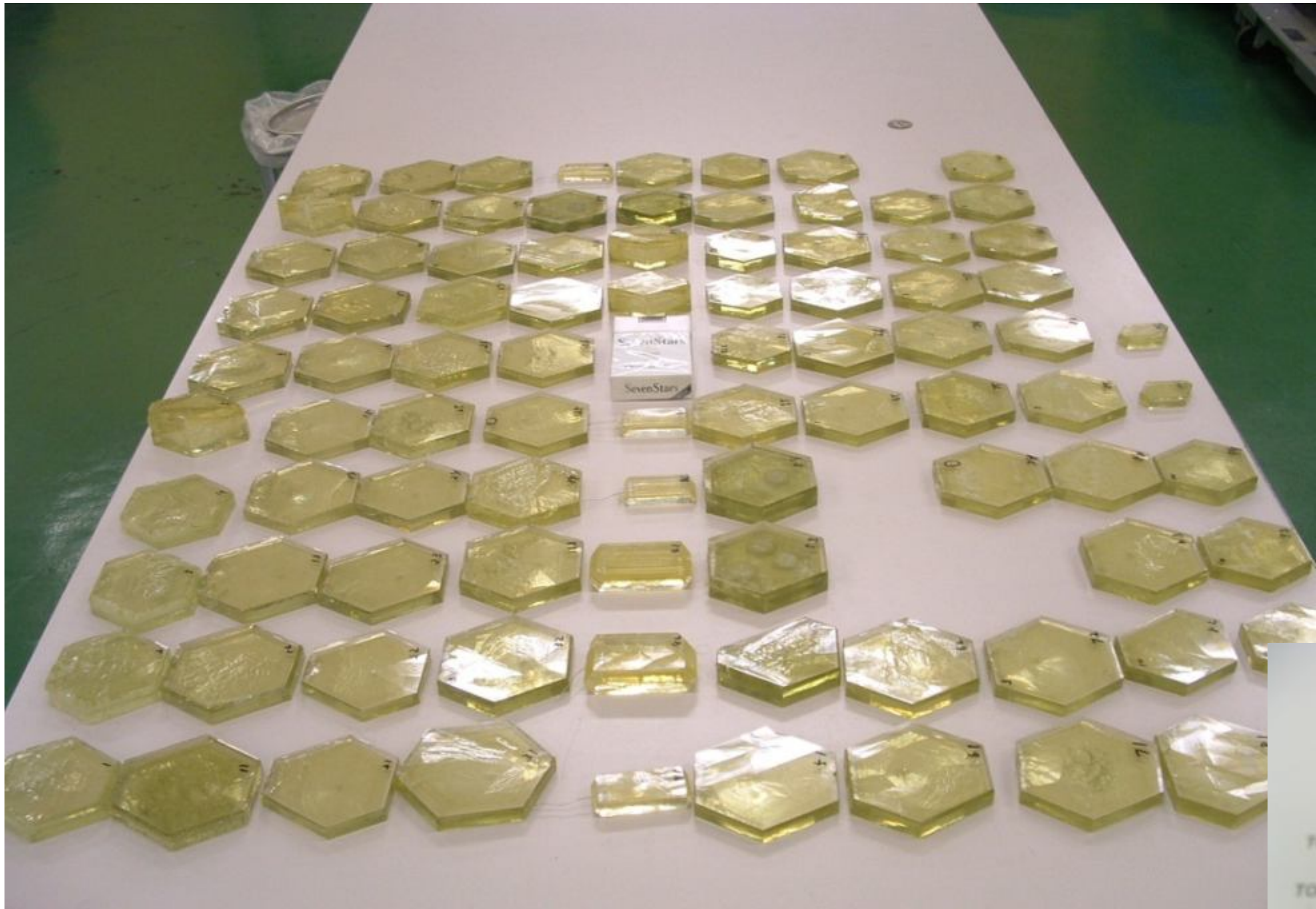


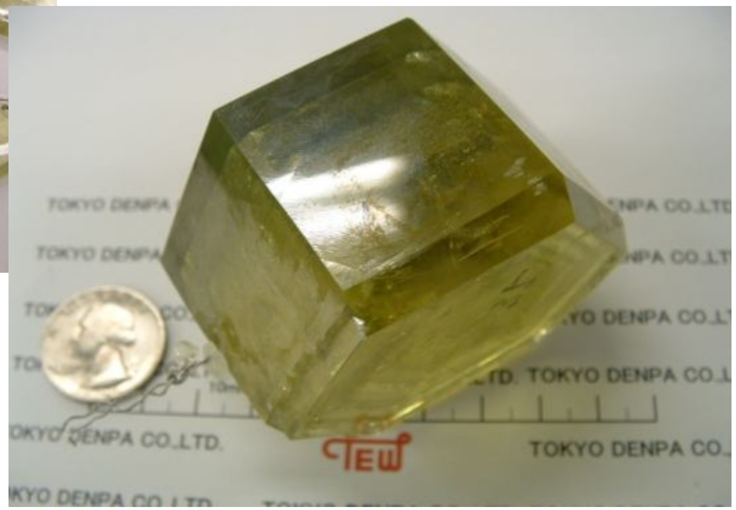
ZnO crystal grown by hydrothermal method



E. Ohshima, et.al. J. Crystal Growth **260** (2004) 166

Large-sized ZnO crystals





APPLIED PHYSICS LETTERS **91**, 231117 (2007)

Hydrothermal method grown large-sized zinc oxide single crystal as fast scintillator for future extreme ultraviolet lithography

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The scintillation properties of a hydrothermal method grown zinc oxide (ZnO) crystal are evaluated for extreme ultraviolet (EUV) laser excitation at 13.9 nm wavelength. The exciton emission lifetime at around 380 nm is determined to be 1.1 ns, almost identical to ultraviolet laser excitation cases. This fast response time is sufficiently short for characterizing EUV lithography light sources having a few nanoseconds duration. The availability of large size ZnO crystal up to 3 in. is quite attractive for future lithography and imaging applications.
[DOI: 10.1063/1.2815920]

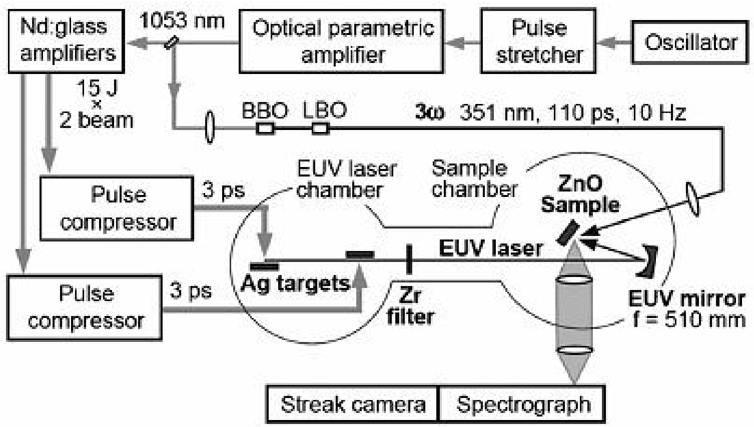


FIG. 1. Experimental setup for the measurement of time resolved spectrum with UV and EUV excitation.

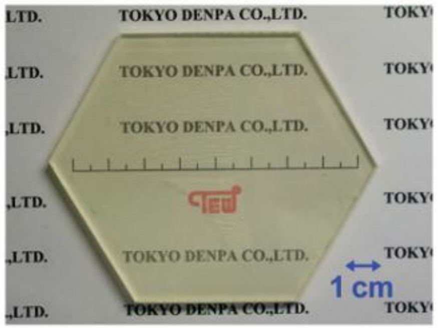


FIG. 3. (Color) Photograph of a 7.5 cm size ZnO crystal grown by hydrothermal method.

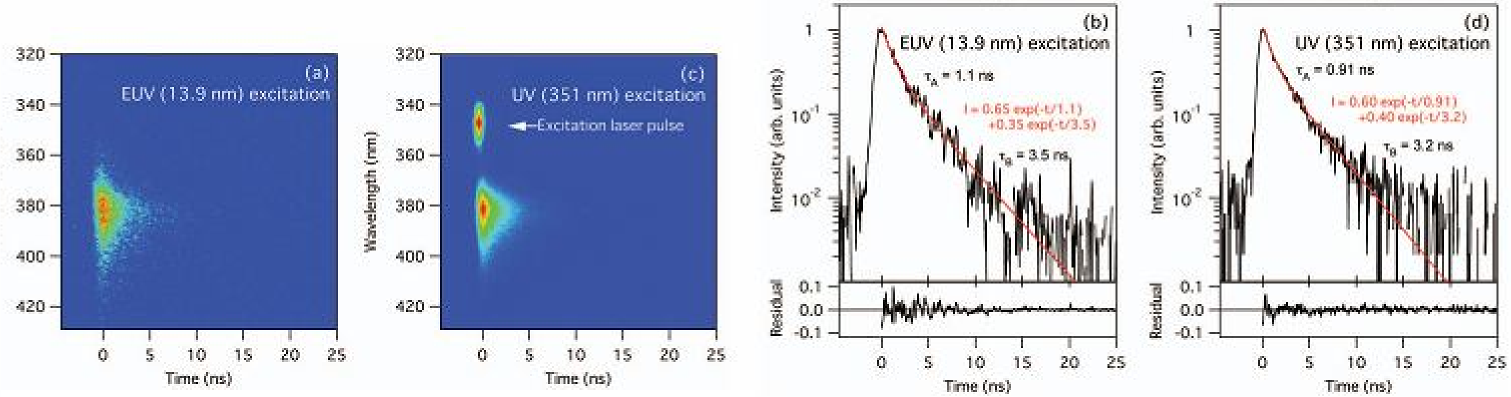
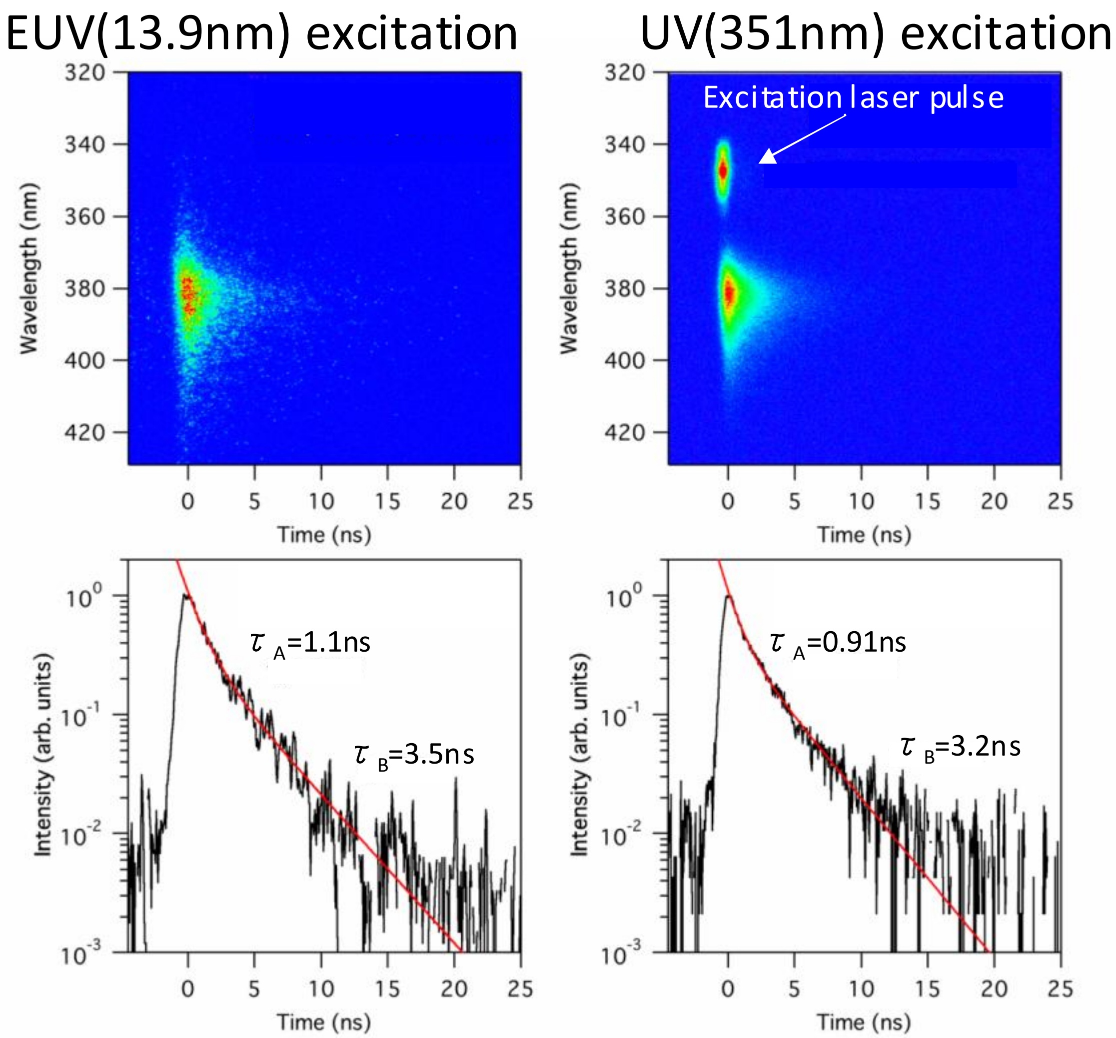


FIG. 5. (Color) Streak camera images and temporal profiles of the fluorescence from ZnO, which are excited by [(a) and (b)] EUV laser pulses (13.9 nm) and [(c) and (d)] UV laser pulses (351 nm). The temporal resolution of this measurement is 0.8 ns. The red lines in (b) and (d) are fitting functions. The observed profiles can be fitted by double exponential decays described as $I = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2)$, as reported in Ref. 8. The fitting parameters for EUV excitation are $A_1 = 0.65$, $A_2 = 0.35$, $\tau_1 = 1.1$ ns, and $\tau_2 = 3.5$ ns, and for UV excitation are $A_1 = 0.60$, $A_2 = 0.40$, $\tau_1 = 0.9$ ns, and $\tau_2 = 3.2$ ns. The time constant and ratio of the two decay components are almost identical in both the excitation conditions.

Fluorescence properties of Conventional ZnO crystal



Tanaka et.al., APL. 91, 231117 (2007)

Double exponential decay τ_A = 1 ns, τ_B = 3 ns

The fluorescence behavior is similar in both cases.

ZnO crystal is a promising scintillation material.

Need to improve response time

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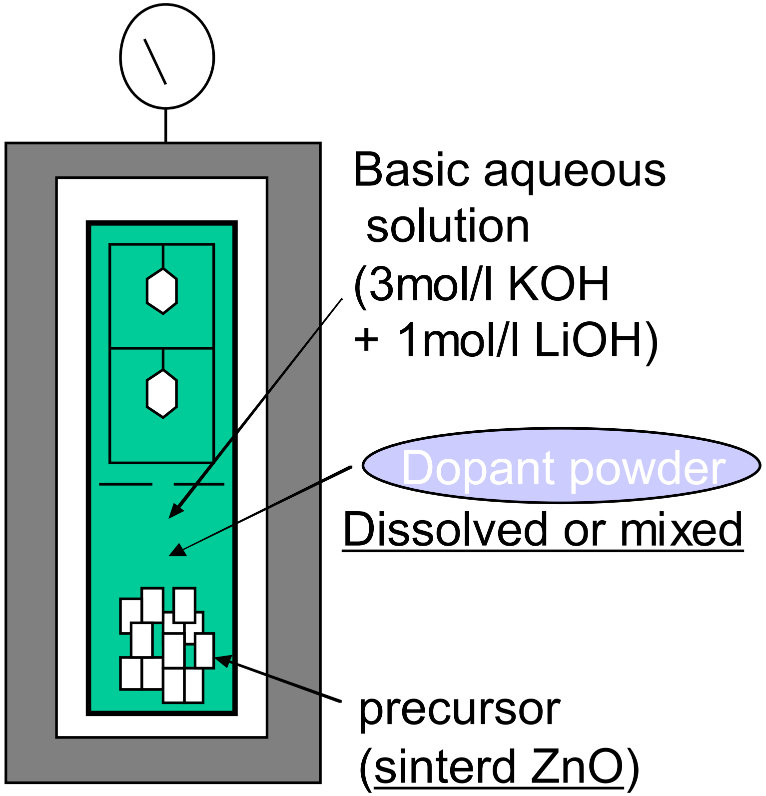
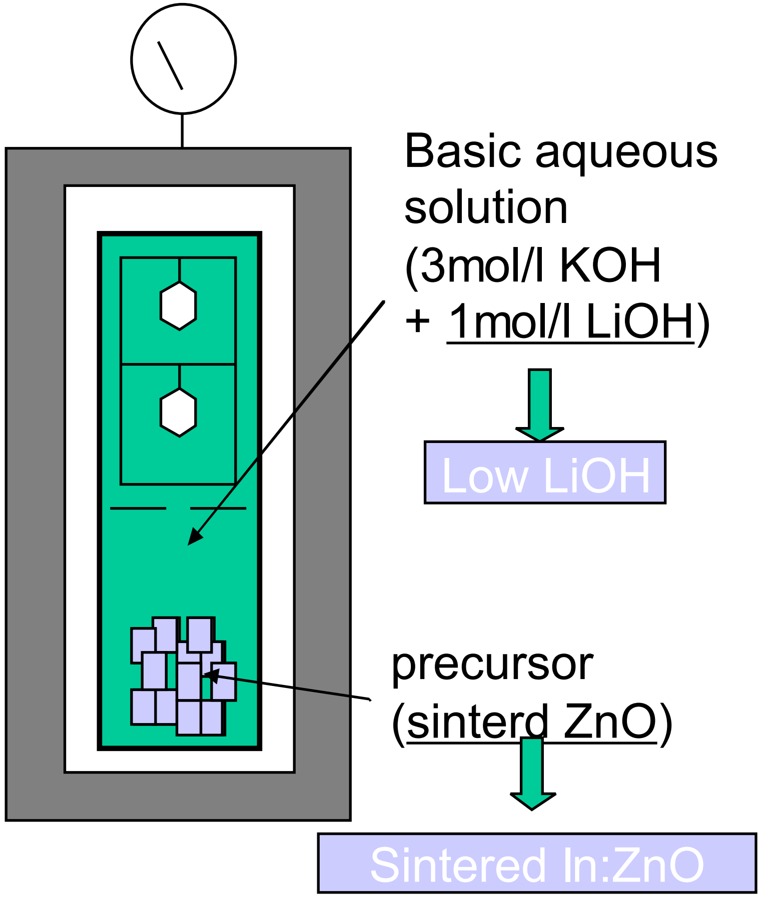
Impurity doping by the Hydrothermal method



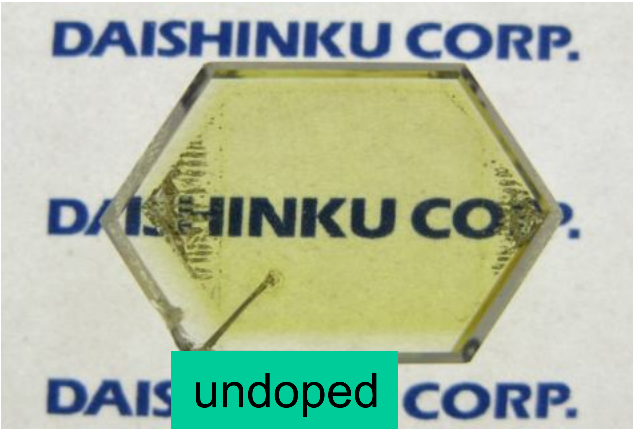
Doping method 1



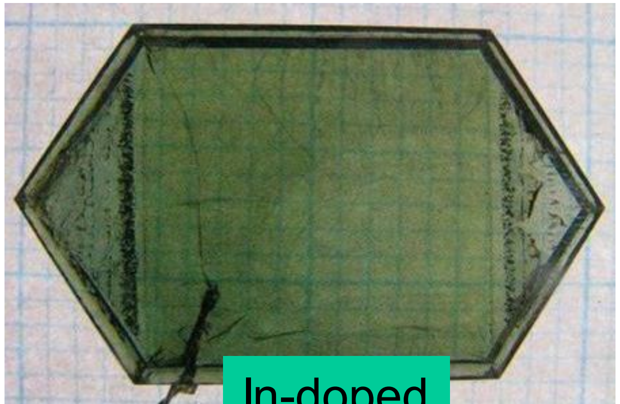
Doping method 2

E. Ohsima et al., J. Cryst. Growth, **260**,166 (2004)

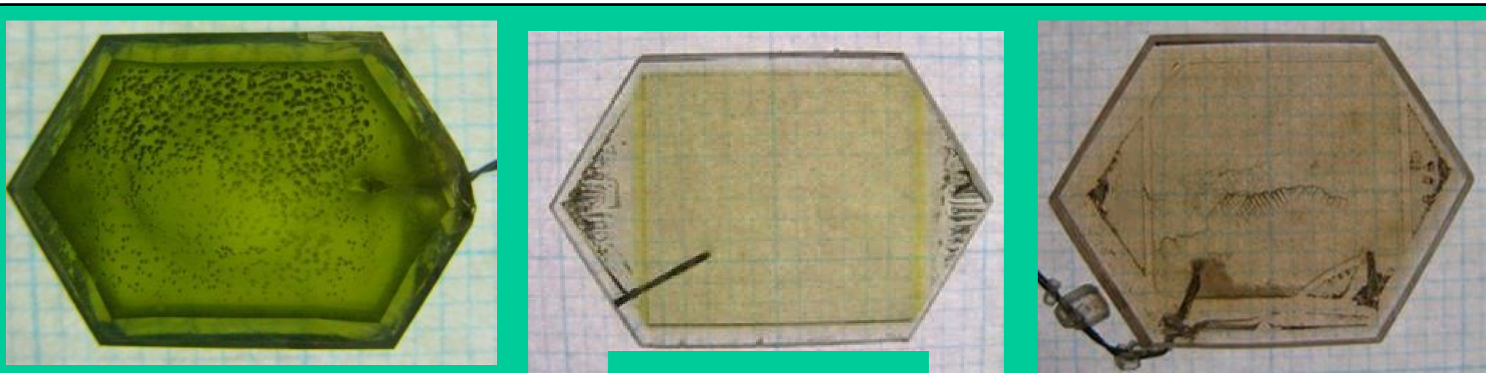
ZnO crystals



undoped

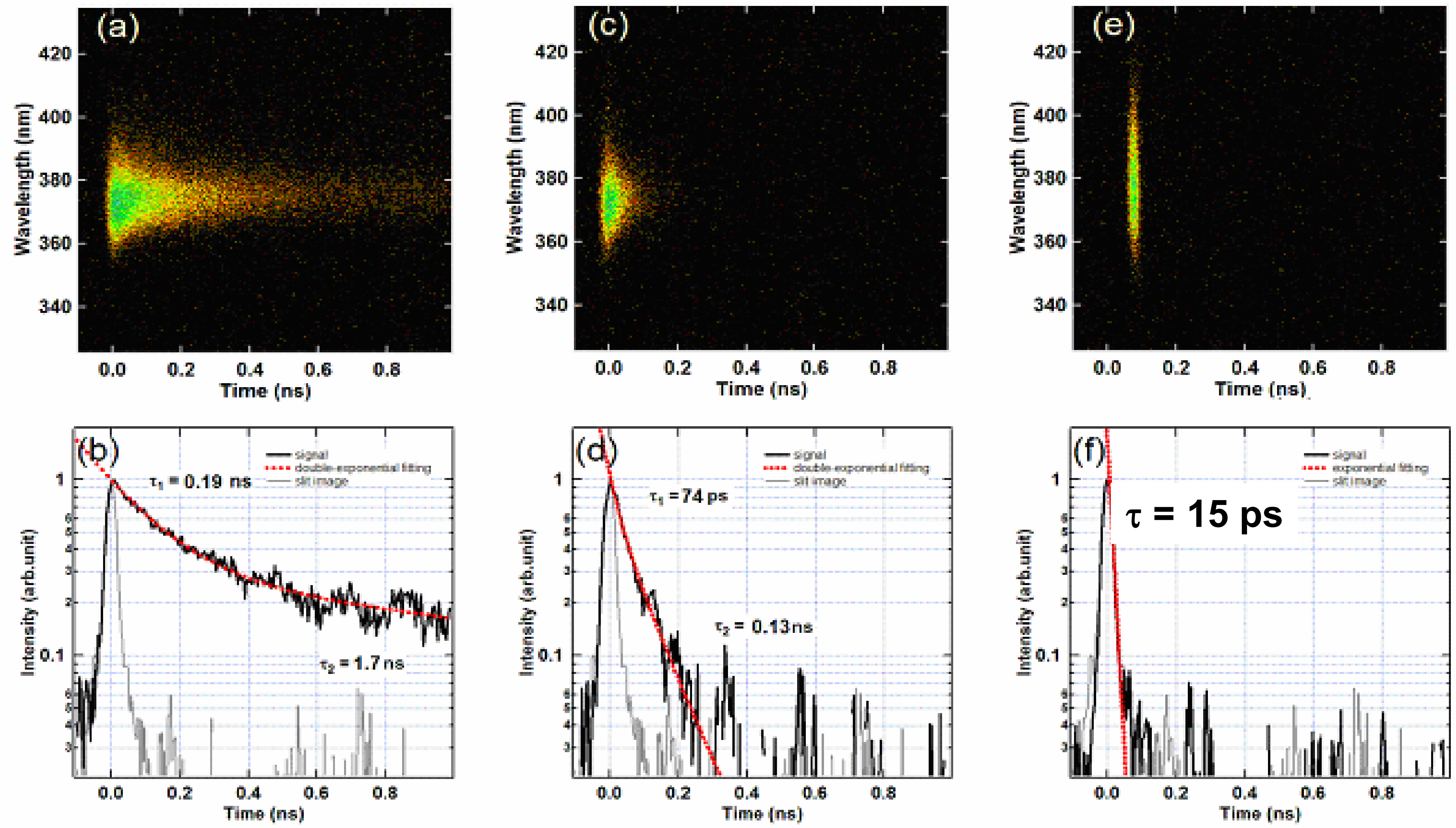


In-doped



Other dopant

Result of lifetime measurements



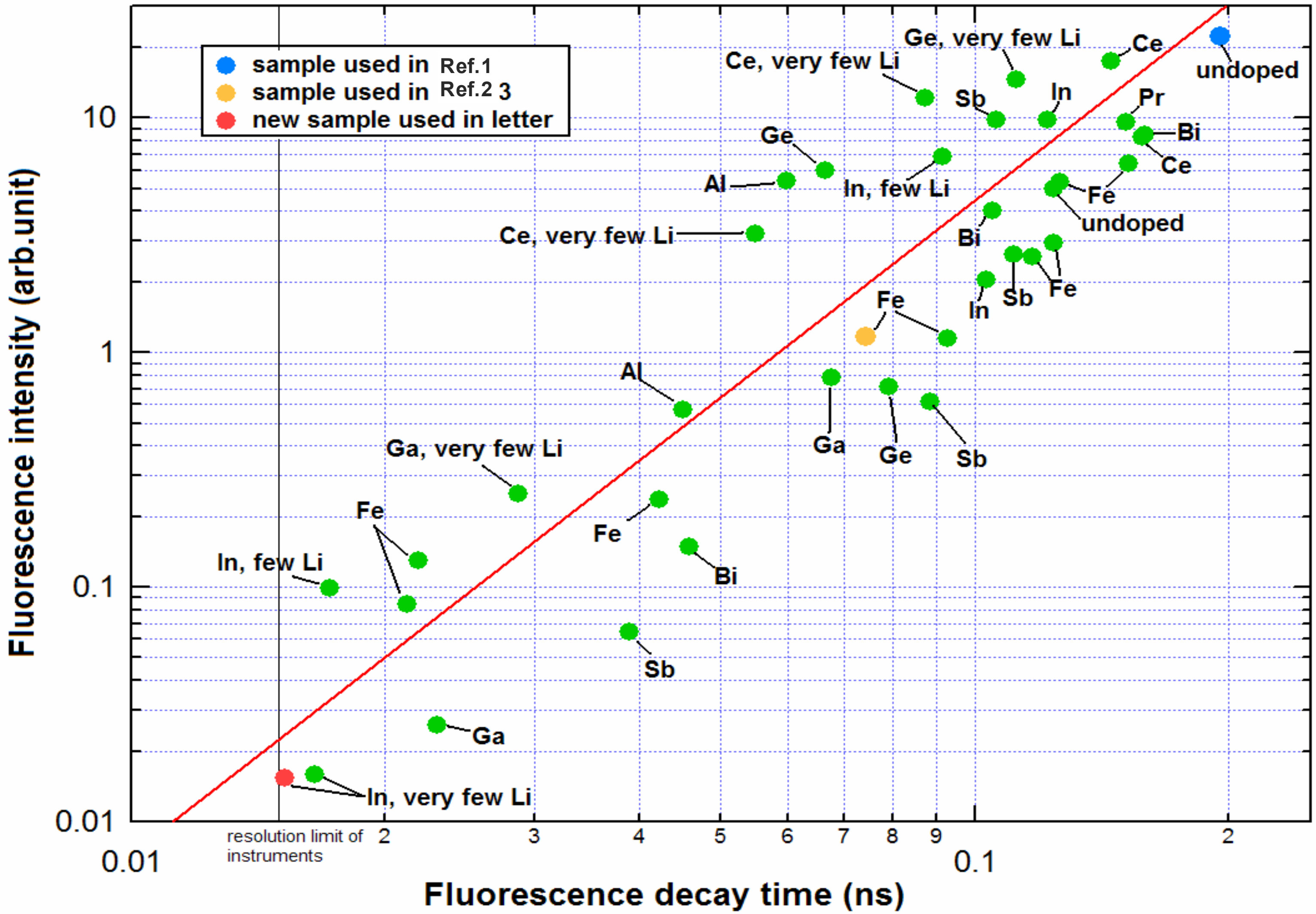
undoped

Fe-doped

In-doped with few LiOH

Yamanoi et.al. Opt. Mat. **32** (2010) 1305–1308

Result of lifetime and light yield measurements



Ref.1 : Tanaka et.al., APL. 91, 231117 (2007)
Ref.2 : Shimizu et.al.,RSI. 81, 033102 (2010)

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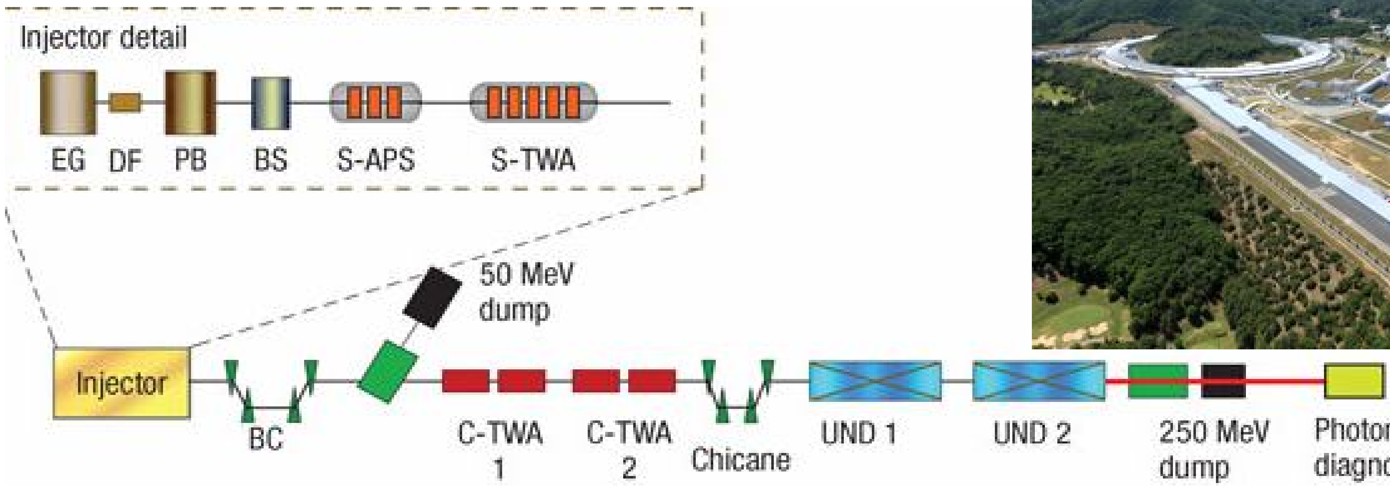
SCSS (SPring-8 Compact SASE Source) test accelerator



SACLA

T. Shintake, et al., Nature Photonics **2**, 555 (2008)

CeB6 Cathode Emitter

A high-quality electron beam is launched from a CeB6 electron gun.

C-band
Accelerating
Structure

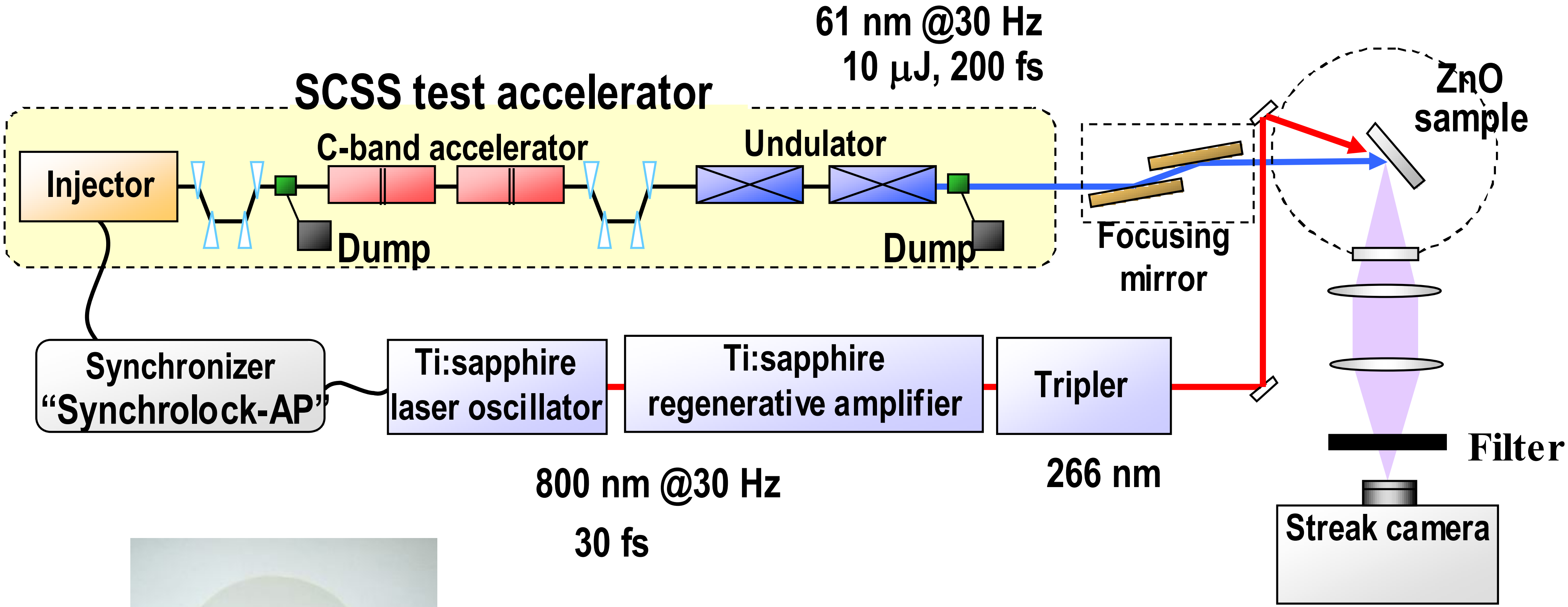
The C-band accelerator swiftly increases the electron energy.

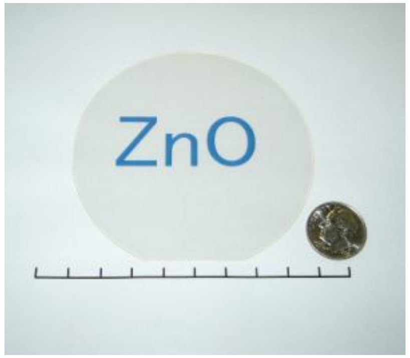
In-vacuum
Undulator

The in-vacuum undulator generates strong XFEL radiation.

from the pamphlet of XFEL at SPring-8 (2008)

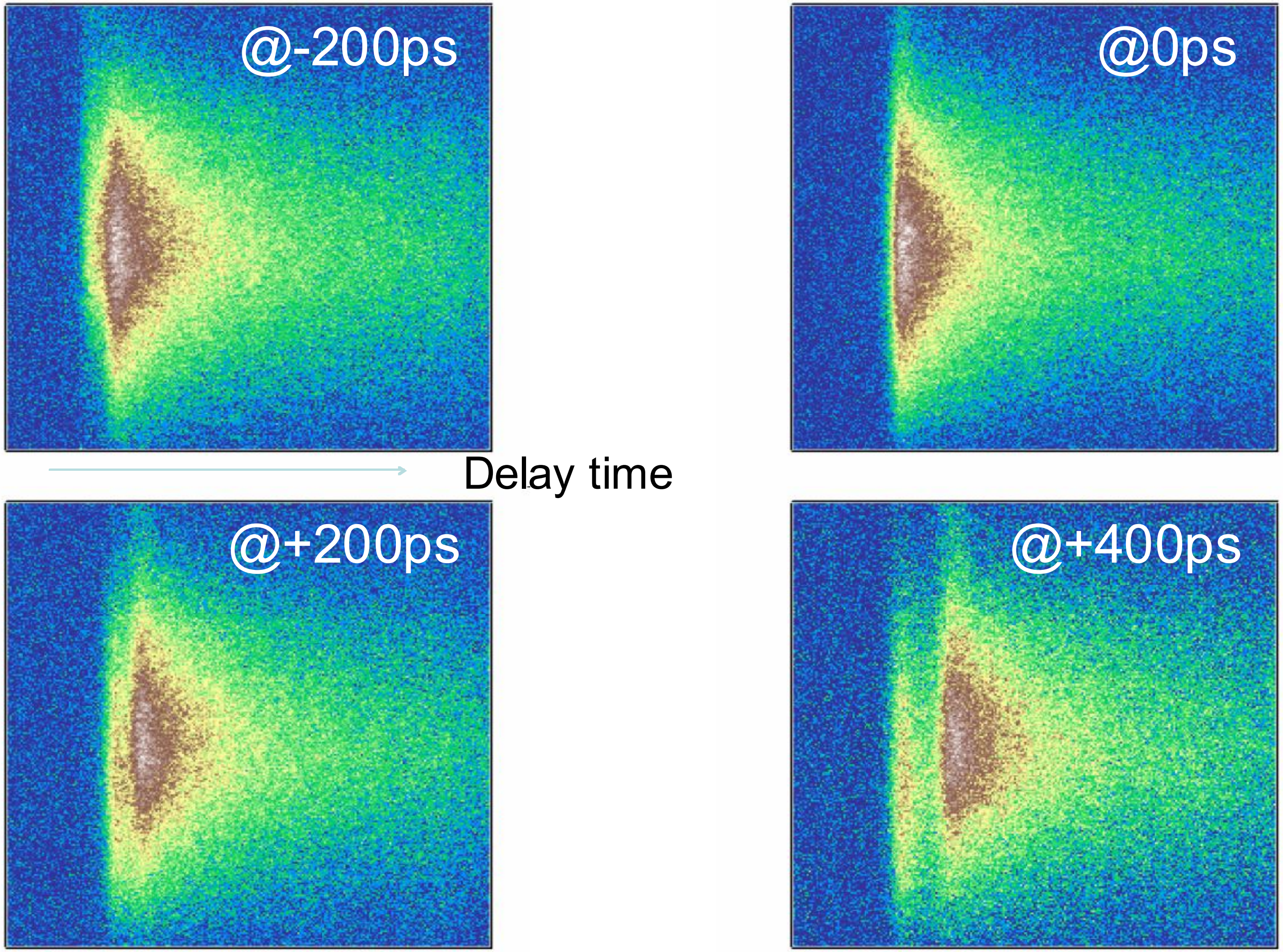
Experimental Setup



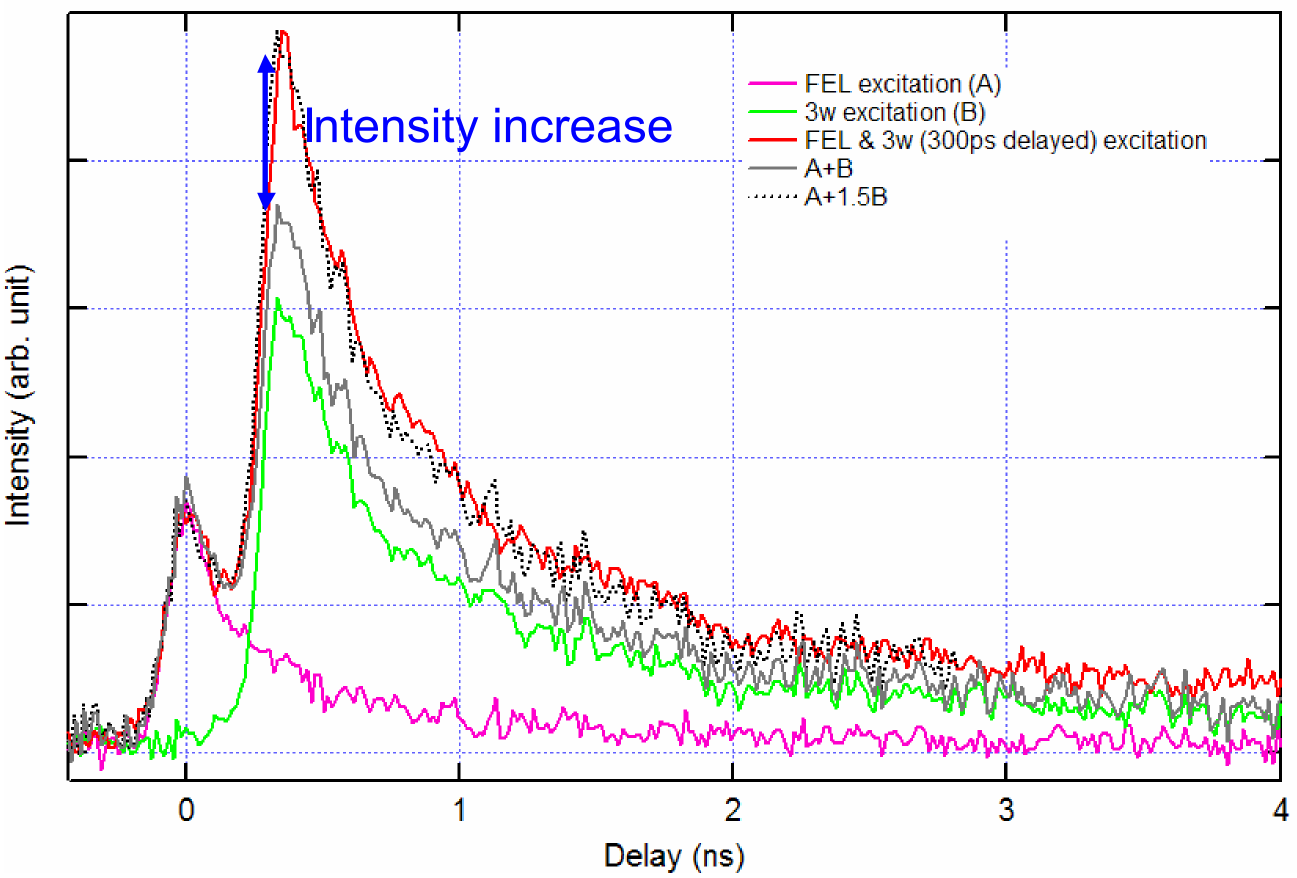


Hydrothermal method grown ZnO crystal

Streak images for different delay times



Analysis: dependence of amplification on delay time



—FEL excitation S_f

—266nm(UV) excitation S_{266}

—Simultaneous irradiation S_{all}

—$S_f + S_{266}$

··· Fitting S_{fit}

$S_{fit} = A \times S_f + B \times S_{266}$

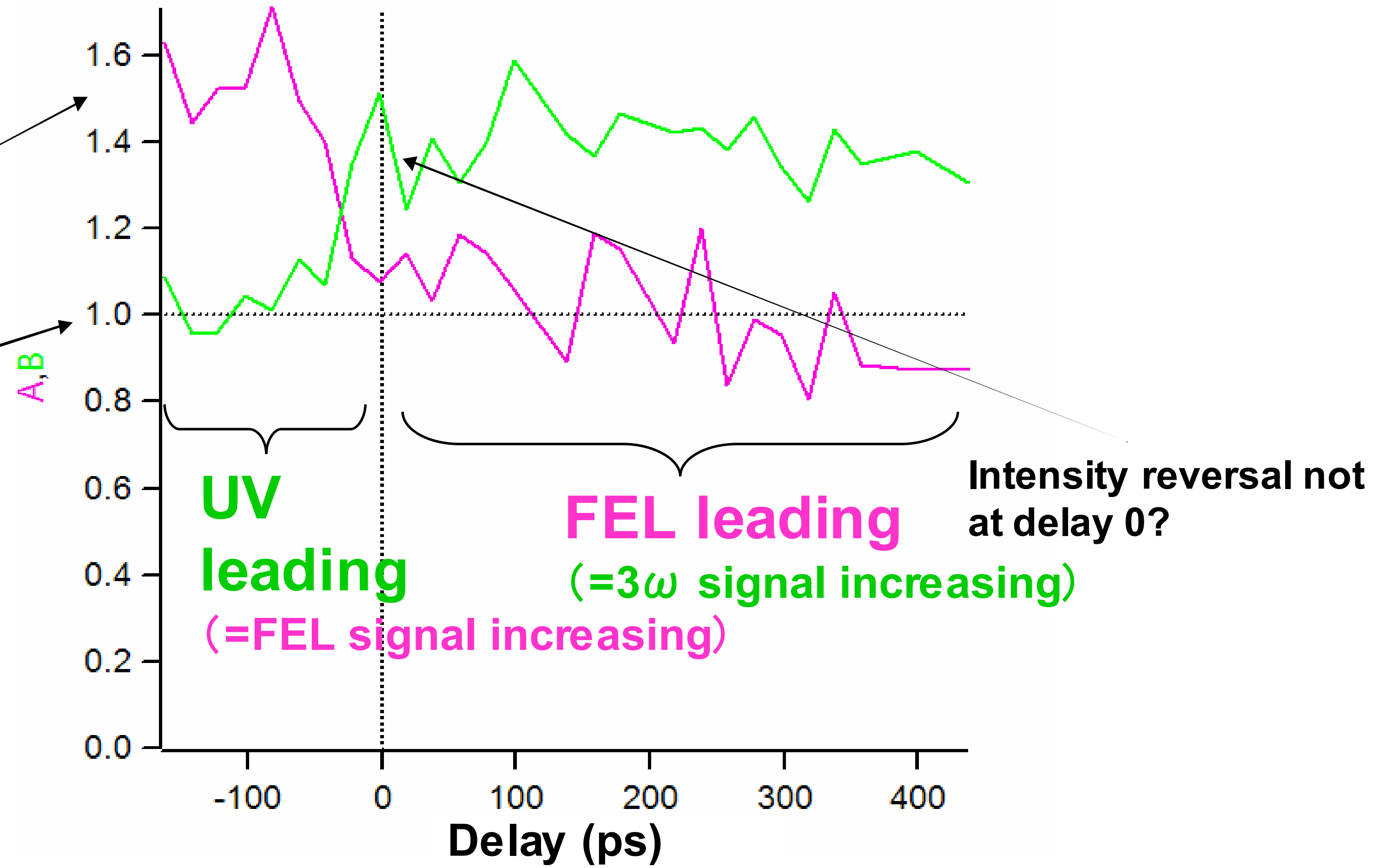
Fitting parameters A and B were defined as S_f multiplied by A and S_{266} multiplied by B, and are equal to S_{all}.

Results: dependence of amplification on delay time

A,B as a function of delay time

Delayed signal amplified up to 1.4 times

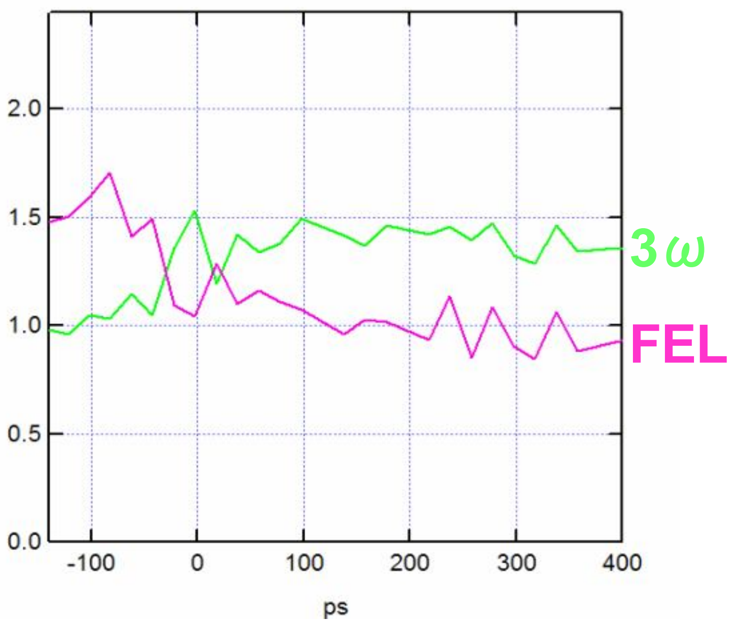
Leading signal not amplified



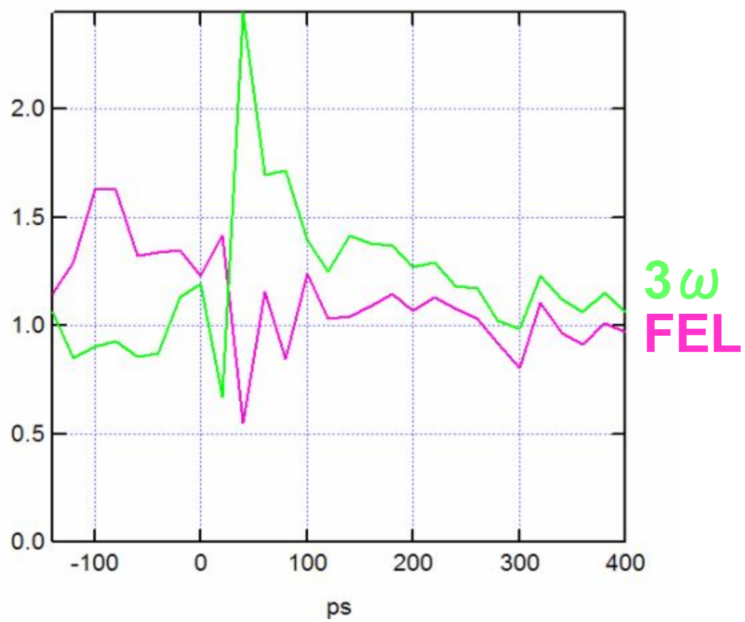
Delayed signal was amplified with a longer decay time

Comparison between undoped and Fe-doped ZnO

undoped



Fe-doped

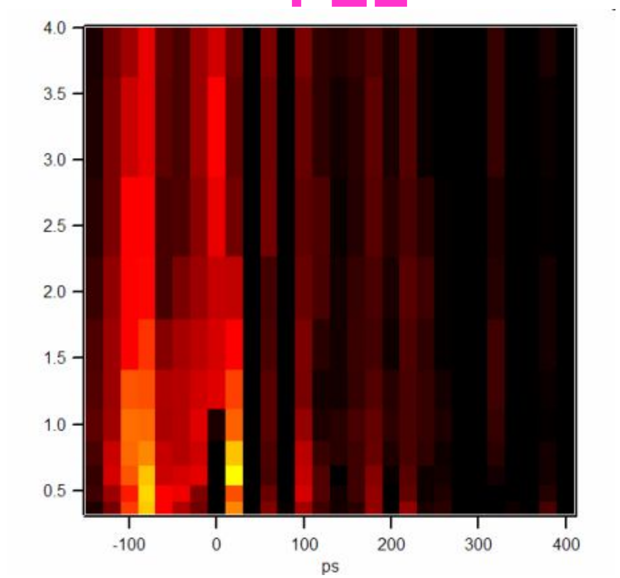


Delay time dependence of amplification is different for undoped and Fe-doped ZnO.

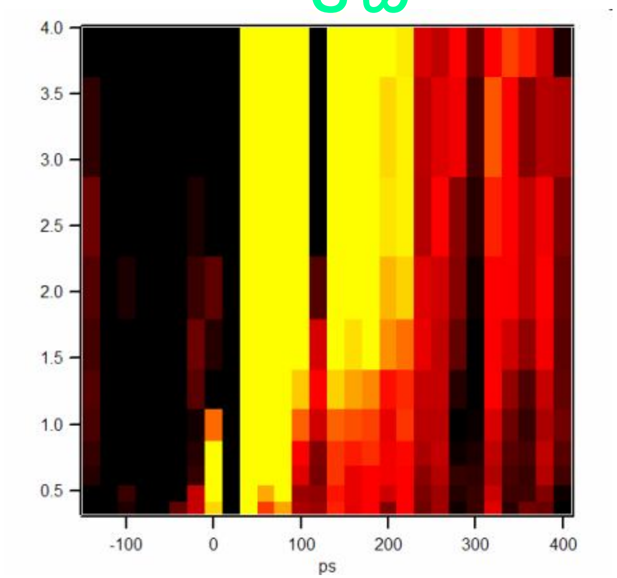
This could be related to the emission lifetime.

Fe-doped ZnO

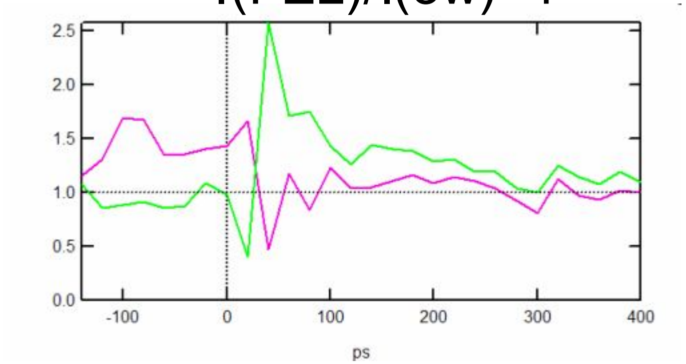
FEL



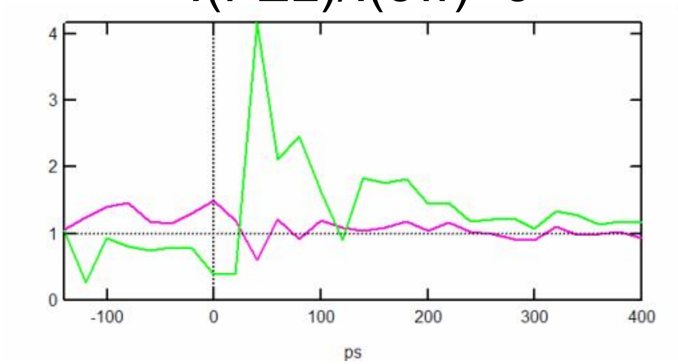
3ω



I(FEL)/I(3w)~1



I(FEL)/I(3w)~3

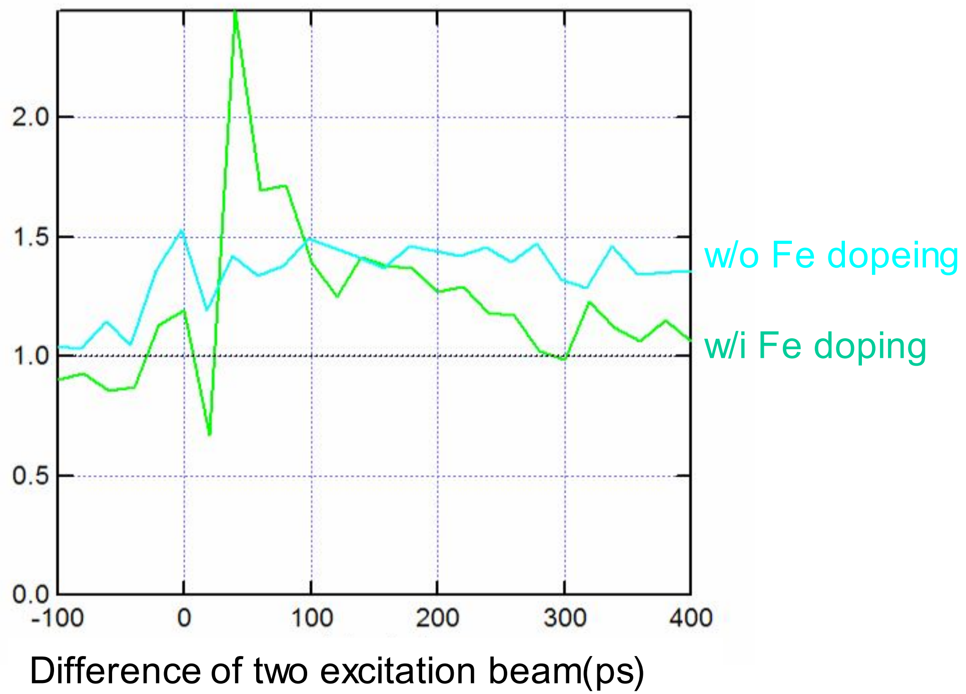


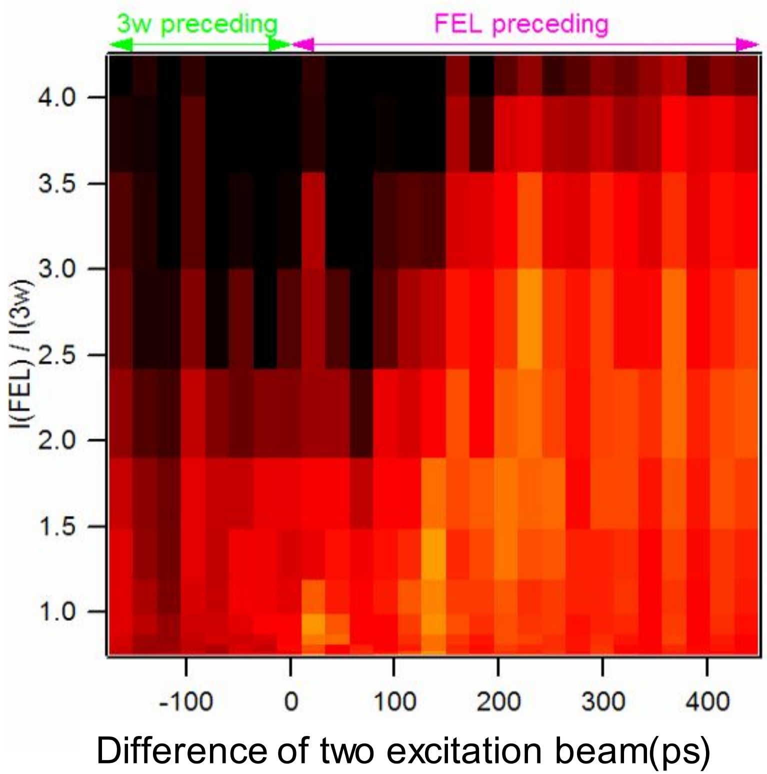
Difference in delay time dependence of amplification for different intensity ratio is not clear.

（Larger FEL intensity, larger amplification?）

Luminescence intensity of undoped ZnO for different intensity ratios between pump and probe light

Delay time dependence of 3ω excitation luminescence

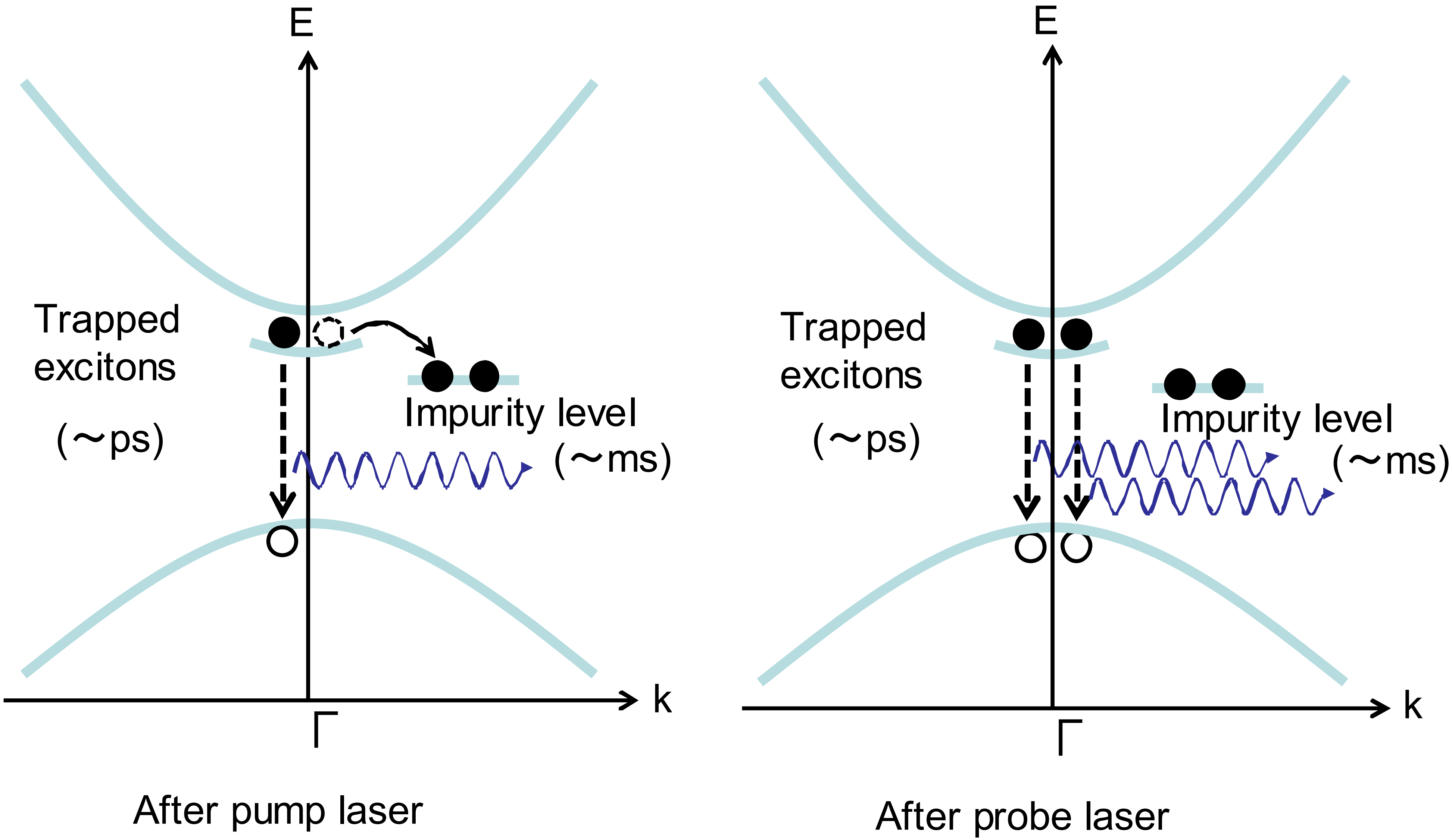




Luminescence originating from 3ω excitation was amplified by FEL excitation. This phenomena depend on the delay time in pump and probe light and the intensity ratio of the two excitation sources.

Hypothesis: Overflow in trap level by intense excitation?

Mechanism of amplification influenced by delay time



Summary

- Pump and probe experiment for ZnO crystal was performed using EUV-FEL（first experiment for solid material）
- Luminescence intensity depends on delay time between pump and probe laser.
- Amplification of luminescence originating from probe light was observed.
- Procedure of pump and probe experiment with FEL was established.

- Future plan: VUV wavelength-emitting material